Development of an Improved Assay for the Determination of Gross Alpha and Beta Concentrations in Soil — Liquid Scintillation Counting

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As a consequence of the nuclear weapons programs, the U.S. Department of Energy (DOE) has approximately 4000 sites covering many thousands of acres that are contaminated with radioactive materials. It is estimated that more than one million samples will need to be analyzed per year in order to support environmental restoration and waste management activities at these sites. In many cases it is unknown what radionuclides may be present, so requests are often made for an isotopic analysis of nuclides from the following groups: actinide elements (Th, U, Np, Pu, Am, Cm); fission products (Sr, Cs, I, Tc); decay products (Ra, Pb); and "gross" alpha and beta activities. If each of these analyses are approached by a commercial laboratory in the traditional manner, a total of 14 analyses would be charged at a typical cost of about $200 each, or a grand total approaching $3 billion dollars per year! The time and cost commitments here are clearly inefficient for the knowledge gained. In addition, typical analysis times are very long, often days to weeks before results are in hand.

What is required is a reliable and complete screening method with low detection limits. In this way samples can be screened and only those that display evidence for the presence of radionuclides above natural levels would be submitted for comprehensive analysis. It is critical that the methods be robust enough that there is a very low probability of anything being missed.

The primary objective of this research is to develop analytical methods for the reliable determination of gross alpha and beta activities in soil samples. We are pursuing this objective by independent and simultaneous evaluation of two standard counting techniques: (1) gas flow proportional counting; and (2) liquid scintillation counting. Both methods are appropriate since they allow for simultaneous alpha/beta counting. On the other hand, both methods may have important limitations which must be taken into account in order to achieve the desired results. We present here a summary of the LSC approach and a separate presentation will cover the direct counting method.

One of the prime benefits of liquid scintillation counting (LSC) is the very high counting efficiency for beta particles and especially alphas at close to 100%. Unfortunately, there are a number of factors which influence count rates and spectral distribution which are not directly related to nuclide abundance and energies. The amount of quenching, for example, is dependent upon the chemical composition of the sample and other variables which are difficult, if not impossible, to control. Our approach to the analysis of total alpha/beta in soil samples is to

upon the chemical composition of the sample and other variables which are difficult, if not impossible, to control when dealing with environmental samples. Our approach to the analysis of total alpha/beta in soil samples is to evaluate experimentally the optimum conditions for running digested soil samples added directly to a high efficiency scintillator (eg. Packard Ultima Gold A/B).

The protocol we are following for sample preparation is based on the digestion of approximately 0.75 g of dried and homogenized soil in teflon microwave digestion bombs with 10 mL of concentrated nitric acid. The microwave unit is programmed to cycle on/off at full power (660 watts) to maintain the pressure below 120 psi over a 30-minute period. After an appropriate cool off period, the resulting solution is filtered through a 0.45 micron filter and the filtered solution is saved in a beaker. The beaker is placed on a hot plate and the solution is brought down to near dryness. After the beaker cools, 5 mL of 8M nitric acid and 1 mL of 30% hydrogen peroxide are added to wet ash the residue. This mixture is evaporated down to dryness without baking the sample. The residue is re-dissolved in 4 mL of 1M nitric acid by shaking and swirling the mixture. The 4 mL of sample is transferred into a clean liquid scintillation vial and 16 mL of Ultima Gold AB cocktail is added to the vial and the cap is sealed tightly onto the vial. Vials are shaken well and allowed to dark adapt for at least one hour before counting.

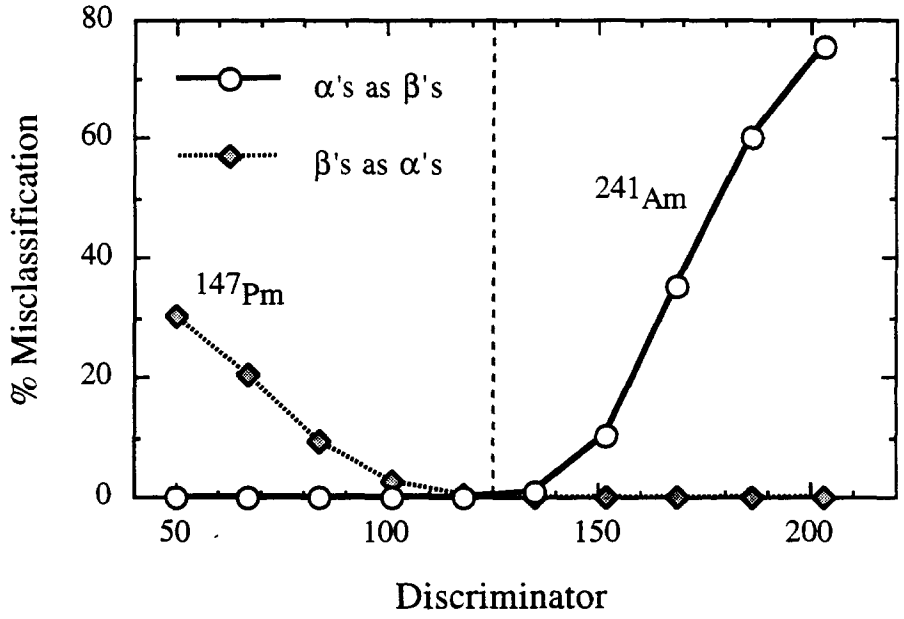

Figure 1. Percent misclassification versus discriminator setting plot obtained by running a pure α-particle emitter (^{241}Am) and a pure β-emitter (^{147}Pm). The optimum setting under these conditions was determined to be 124.

We set the conditions for simultaneous alpha/beta counting on a Packard 2550 TR/AB by first running pure alpha (^{241}Am) and pure beta (^{147}Pm) standards at different discriminator settings to evaluate the optimum setting at one quench level (**Fig. 1**). Since the percent misclassification depends on both the discriminator setting and level of quench (measured as tSIE), it is necessary to adjust the corrections for the observed quench. We evaluated this by developing a percent misclassification versus tSIE curve (**Fig. 2**) at one discriminator setting by

running a series of spiked samples at different mass loadings that span the range of quench factors [tSIE factors ≈ 260 - 385] observed in the majority of our soil samples prepared by the microwave digestion protocol.

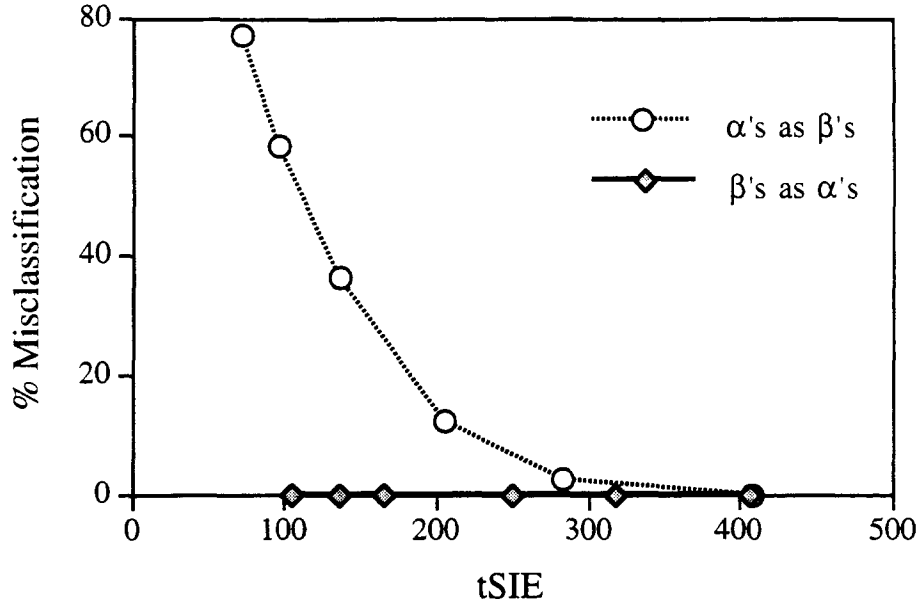

Figure 2. Percent misclassification versus quench level (expressed as tSIE) determining by running a series of spiked samples with different amounts of soil.

Our initial results suggest that we can reliably obtain an MDA for "total" alphas of <5 pCi/g in about a 10-minute counting period. Furthermore, a counting error of <10% should be achievable with a counting time of only about 15 minutes for a typical total alpha activity of approximately 15 pCi/g. It should be noted that this approach may not provide a pure representation of the actual "total" alpha or beta activity since it is based on a digestion, rather than a complete dissolution, of the soil samples. However, in many cases the strong acid soluble fraction should provide the necessary information and, after further refinement, this method should easily fit into a monitoring program to screen samples for total alpha and beta activities.

[#] For presentation at the 42*nd* Annual Conference on Bioassay, Analytical and Environmental Radiochemistry; San Francisco, October 13-17, 1996.

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